

Trends in Concentrations of Polychlorinated Biphenyls in Fish Tissue from Selected Sites in the Delaware River Basin in New Jersey, New York, and Pennsylvania, 1969-98

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Polychlorinated biphenyl (PCB) concentrations in fish tissue collected during the 1990's from selected sites in the Delaware River Basin were compared with concentrations in fish tissue collected during 1969-88. Data collected by State and Federal agencies on concentrations in whole-body common carp (*Cyprinus carpio*) and white sucker (*Catostomus commersoni*), and edible portions of American eel (*Anguilla rostrata*), smallmouth bass (*Micropterus dolomieu*), and channel catfish (*Ictalurus punctatus*) during 1969-98 were compiled to define temporal trends in concentrations of PCBs in fish tissue from selected segments of the Delaware River, Lehigh River, Schuylkill River, and Brandywine Creek.

The Delaware River in the vicinity of Trenton, New Jersey and Yardley, Pennsylvania (above the tidal influence) had the largest long-term data set among the sites considered for this study and was the only site with sufficient data for statistical analysis. A general pattern of decline in PCB concentrations during 1969-98 was apparent for this river segment. PCB concentrations in whole-body white sucker from this lower Delaware River segment declined during 1969-98 from a highest concentration of 7 micrograms per gram ($\mu\text{g/g}$, wet weight) in a sample collected during 1972 to 0.26 $\mu\text{g/g}$ (wet weight) in a sample collected during 1998. PCB concentration was negatively correlated with year (Spearman rank correlation -0.46, $p < 0.08$, $n = 15$); especially after removal of a sample from 1977 with an unusually low concentration (Spearman rank correlation -0.53, $p = 0.05$, $n = 14$). PCB concentrations in edible flesh of American eel declined during 1975-95, from a highest concentration of 3.8 $\mu\text{g/g}$ (wet weight) in a sample collected during 1976 to less than the reporting limit of 0.26 $\mu\text{g/g}$ (wet weight) in samples collected during 1993 and 1995. PCB concentrations in most samples (for species considered in this study) collected from the lower Delaware River exceeded the National Academy of Sciences and National Academy of Engineering (NAS/NAE) wildlife guideline level of 0.5 $\mu\text{g/g}$ during the 1970's and 1980's, and decreased to below this level during the 1990's. No samples of edible portions of game fish exceeded the U.S. Food and Drug Administration (FDA) tolerance level by the mid 1980's. However, the PCB concentration in a smallmouth bass fillet sample that was collected during 1998 (0.37 $\mu\text{g/g}$) exceeded the Pennsylvania fish-consumption advisory level of 0.06 $\mu\text{g/g}$, and the concentrations in whole-body common carp and white sucker collected during 1998 (1.10 $\mu\text{g/g}$ and 0.26 $\mu\text{g/g}$, respectively) exceeded the New York State Department of Environmental Conservation wildlife criterion concentration of 0.11 $\mu\text{g/g}$. (The concentration in carp also exceeded the 1973 NAS/NAE wildlife guideline concentration of 0.5 $\mu\text{g/g}$.)

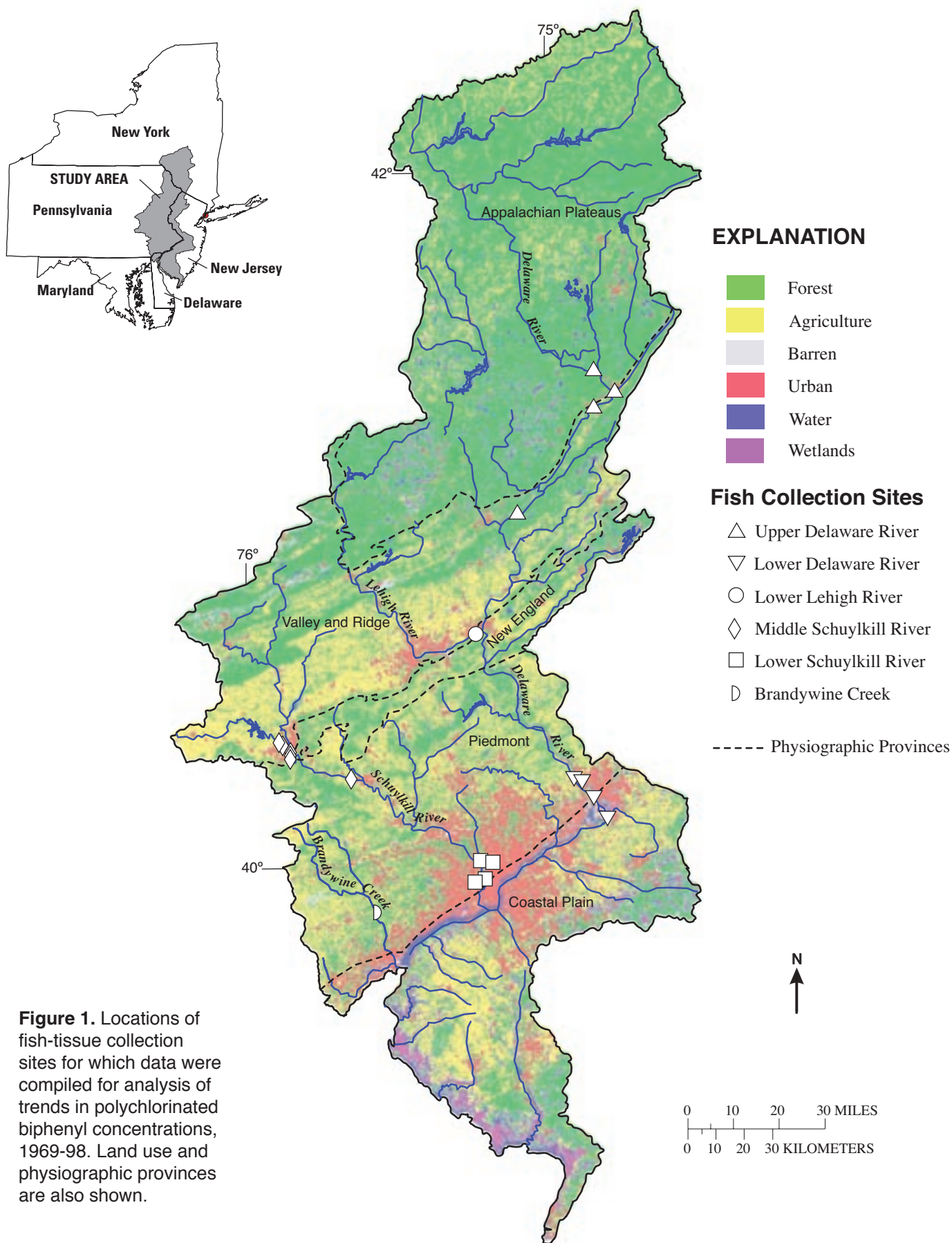
Graphical analysis of PCB concentrations in whole white sucker and (or) edible portions of American eel from the upper Delaware River, lower Delaware River, middle Schuylkill River, and Brandywine Creek indicate a decline from the 1970's and (or) 1980's to the middle to late 1990's. Temporal trends in PCB concentrations in white sucker samples from the lower Lehigh and Schuylkill Rivers during 1979-98 are less

clear; the PCB concentration (wet-weight basis) from a sample collected in 1998 from the lower Lehigh River was similar to that from a sample collected in 1979, and concentrations actually increased during 1982-98. Similarly, PCB concentrations in samples of white sucker and American eel from the lower Schuylkill River were highly variable over time. A decrease in lipid-adjusted PCB concentrations at both sites (for several white sucker samples with available lipid data) indicates that differences in fat content could be masking actual declines at these sites, but the data are insufficient for further interpretation. The available data indicate a slower rate of decline in PCB concentrations in fish from the lower Lehigh and Schuylkill Rivers than in fish from the other rivers considered in this study.

The highest PCB concentrations in whole-body white sucker and common carp collected during 1998 from the study sites were in specimens from the lower Schuylkill River. These concentrations (4.0 $\mu\text{g/g}$ and 0.89 $\mu\text{g/g}$, respectively) exceeded the national geometric mean concentration for whole-body fish in 1979 reported by the National Contaminant Biomonitoring Program. Recent (1994-1998) site-to-site differences in PCB concentrations in fish tissue, and in the patterns of decline since the 1970's and 1980's, correspond with amounts of urban-industrial land, population density, and point sources within watersheds, and may also be related to the retention, resuspension, and movement of PCB-contaminated sediment from a variety of sources. Most sites studied indicate a decline in fish tissue PCB concentrations since the 1970's and 1980's, but concentrations in certain species at some sites remain sufficiently high by the mid- to late- 1990's to pose concern for human and wildlife health.

INTRODUCTION

Polychlorinated biphenyls (PCBs) were banned throughout the United States in 1979 but continue to be detected in streambed sediments and fish tissue collected from streams and rivers of the Delaware River Basin (fig. 1), as in much of the rest of the Nation. Concentrations in fish tissue have remained sufficiently high in some streams and rivers of the Delaware River Basin to pose a risk to humans. This risk has prompted State consumption advisories (Pennsylvania Fish and Boat Commission, 2000; New Jersey Department of Environmental Protection, 2000). Fish-eating wildlife



such as mink, otter, osprey, eagles, and predatory fishes also can be harmed by current concentrations of PCBs that exceed criteria or guidelines.

PCBs are organochlorine compounds that were widely used during 1929-74 in many types of chemical and electrical products, including paints, inks, plasticizers, hydraulic fluids, electrical transformers, and capacitors. PCB production in the United States was banned after 1979 (Niimi, 1996; U.S. Environmental Protection Agency, 1992a), but they still are present in some closed electrical systems, streambed and terrestrial sediments, landfills, and other potential sources of contamination to aquatic environments. PCBs sorb to sediments and are relatively water-insoluble and lipophilic, which gives them a tendency to accumulate in the tissues of aquatic organisms, many of which, in turn, are consumed by birds, mammals, and predatory fishes. PCBs are toxic, carcinogenic, and mutagenic (Eisler, 1986; Colborn and others, 1993), and have been implicated in endocrine disruption of fish and wildlife (Colborn and others 1993; U.S. Environmental Protection Agency, 1997).

High concentrations of PCBs were found in fish from certain rivers in the Delaware River Basin more than 20 years ago. For example, in a study conducted during 1976 of 62 sites throughout Pennsylvania, Brezina (1980) found the highest concentrations in the lower Delaware and Schuylkill Rivers. PCB concentrations in edible flesh of American eel (*Anguilla rostrata*) and common carp (*Cyprinus carpio*) collected from four sites in the middle to lower segments of the Schuylkill River, and one site in the most downstream segment of the nontidal Delaware River (fig. 1), ranged from 3.69 $\mu\text{g/g}$ to 7.5 $\mu\text{g/g}$. These concentrations were higher than those found at all other Pennsylvania sites except for one (Brezina, 1980). PCB concentrations in some samples of adult white suckers (*Catostomus commersoni*) collected from the middle section of the Schuylkill River during a 1977 study were even higher than those reported in the 1976 study (median concentration 9.01 $\mu\text{g/g}$; ranging from 0.39 $\mu\text{g/g}$

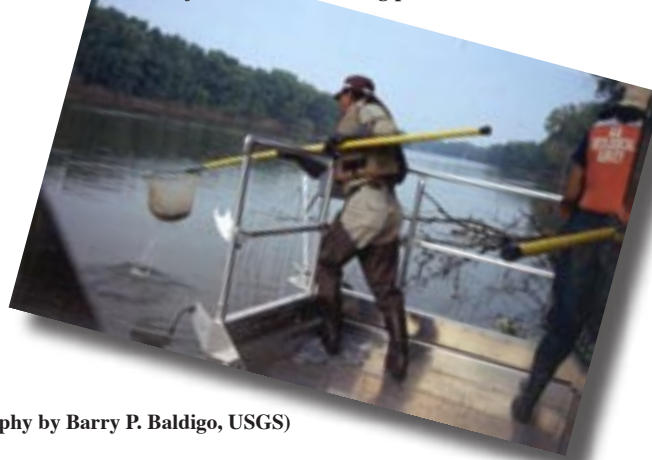
in young fish to 18.88 $\mu\text{g/g}$ in older fish, Brezina, 1980). These river segments have major urban-industrial areas with many potential sources of PCBs in their watersheds. These findings parallel those of a large national study conducted during 1967-1979, which found higher PCB concentrations in fish from rivers in urban-industrial areas of the Northeast and Midwest than in fish from rivers in other land-use settings throughout the Nation (Schmitt and others, 1983). A recent report by the Delaware River Basin Commission (1998) states that streams and rivers flowing into the Delaware Estuary continue to contribute large amounts of PCBs to the estuary, where concentrations in sediments and fish are found at levels thought to be harmful to ecological well-being and human health. A committee of environmental professionals and concerned citizens identified PCB contamination of water, sediment,

and fish as one of the most important water-quality issues of concern in the Delaware River Basin (Fischer, 1999). These concerns were presented to the U.S. Geological Survey (USGS) at planning meetings for the National Water-Quality Assessment Program (NAWQA) study of the Delaware River Basin.

Fish and streambed sediments were collected from 25 sites throughout the Delaware

River Basin during 1998-99, as part of the Delaware River Basin NAWQA study. These samples were analyzed for PCBs, organochlorine pesticides, trace elements, and other contaminants (DeLuca and others, 1999). PCBs and other long-banned organochlorine compounds were detected in fish and sediments from many of these streams and rivers and concentrations at some of these sites were elevated above existing guidelines for human-health and wildlife-health protection. PCBs were detected in whole-body white sucker samples from 21 of the 25 sites sampled, and in common carp samples from all 5 of the sites from which this species was also collected (table 1, DeLuca and others, 1999); detectable concentrations ranged from 0.075 to 4 $\mu\text{g/g}$. Although this study was not a random survey of sites, the detection of PCBs in whole-body fish tissue from a large percentage of sites in forested, agricultural, urban,

"PCB concentrations in fish collected during the 1990's were compared with those in fish collected during previous decades"



(Photography by Barry P. Baldigo, USGS)

and mixed land-use watersheds indicates the persistence and widespread occurrence of PCBs in the Delaware River Basin. These findings prompted an examination of how PCB concentrations in the 1990's compare with those measured during 1969 to 1989 at selected sites in the Delaware River Basin.

PURPOSE AND SCOPE

This report (1) describes the study area and methods, (2) presents recent (1994-98) PCB concentrations for selected fish species and sites and relates these to selected human-health and wildlife-health guidelines or criteria, (3) depicts, in graphical form, the changes in PCB concentrations over time for selected species and sites, and (4) discusses factors that may affect observed site-to-site differences in trends in PCB concentrations.

STUDY AREA

The Delaware River Basin encompasses more than 12,700 mi² in parts of Pennsylvania, New Jersey, New York, and Delaware. The NAWQA study area, depicted in figure 1, includes all of the Delaware River Basin except for about 770 mi² of the Coastal Plain in Delaware and the tidal part of the Delaware Estuary (Fischer, 1999). The two major tributaries of the Delaware River within the study area are the Schuylkill River, which drains 1,893 mi², and the Lehigh River, which drains 1,359 mi². The study area lies within five physiographic provinces (Parker and others, 1964), four of which make up more than 90 percent of the study area. Land use varies widely among the physiographic provinces (fig. 1). The Piedmont and Coastal Plain physiographic provinces contain 80 percent of the study area's population of 7.2 million. About 20 percent of this area is urban land, much of which was agricultural and (or) forested land until recently. The Valley and Ridge physiographic province contains 14 percent of the population and is about 33 percent agricultural land, and the Appalachian Plateaus physiographic province to the north is about 85 percent forested (Fischer, 1999).

PREVIOUS STUDIES

Fish-tissue samples have been collected from the tidal and nontidal parts of the Delaware River and its tributaries for chemical-residue analysis since 1969.

Table 1. Total polychlorinated biphenyl (PCB) concentrations detected in whole white sucker (*Catostomus commersoni*) and common carp (*Cyprinus carpio*) collected during 1998-99, in relation to dominant land use, Delaware River Basin. [$\mu\text{g/g}$, micrograms per gram, wet weight; n, number of samples; -- not collected.]

Land use and species	Median concentration ($\mu\text{g/g}$) ^a	Percentage of sites with PCB detection	n
Urban			
Sucker	0.22	100	8
Carp	.45	100	2
Agricultural			
Sucker	.09	75	8
Carp	--	--	--
Forest			
Sucker	.07	60	5
Carp	--	--	--
Mixedb			
Sucker	.45	100	4
Carp	1.10	100	3

a $\mu\text{g/g}$ = 1 part per million (ppm) = 1000 parts per billion (ppb)

b describes watersheds with mixed agricultural, urban, and forested land cover

The states of Delaware, New Jersey, New York, and Pennsylvania routinely monitor PCB concentrations in a variety of fish species from many sites, primarily to establish consumption advisories for human-health protection. Several rivers and streams in the Delaware River Basin have been included in large national studies that were designed to document national or regional trends and (or) spatial patterns of PCB concentrations. The National Contaminant Biomonitoring Program (NCBP, Schmitt and others, 1999) collected whole-body fish from the Delaware River at Trenton, N.J. during 1969-88; The U.S. Environmental Protection Agency collected fish from the Schuylkill and Lehigh Rivers during 1986-87 as part of their National Study of Chemical Residues in Fish (NSCRS, U.S. Environmental Protection Agency, 1992a,b), and collected various species from some small streams during 1987 for contaminants analysis as part of the Environmental Monitoring and Assessment Program. The USGS NAWQA program collected fish for contaminants analysis from 25 sites in the Delaware River Basin during 1998-99.

NATIONAL WATER-QUALITY ASSESSMENT PROGRAM

The Delaware River Basin is one of more than 50 river basins and groundwater systems across the Nation that are being studied as part of the National Water-Quality Assessment program. NAWQA goals are to (1) provide a consistent description of current water-quality conditions within a large part of the Nation's water resources, (2) define temporal trends in water-resource quality, and (3) identify natural and human factors that affect these conditions and trends. NAWQA's multidisciplinary approach includes surveys of contaminants in fish tissue and streambed sediments, in addition to studies of surface-water and ground-water quality, streamflow, stream geomorphology and habitat, and aquatic communities (Leahy and others, 1990). A committee of environmental professionals and concerned citizens from governmental agencies, private organizations, and educational institutions within the study area provides guidance and information for each NAWQA study.

ACKNOWLEDGMENTS

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METHODS OF DATA COMPILATION AND ANALYSIS

Available data sets were reviewed before compilation to determine which data should be aggregated for analysis. Data were sought from (1) large-scale or long-term studies of fish species that spend all or most of their lives in the nontidal part of the Delaware River Basin, (2) sampling programs with published protocols and data-quality information, and, (3) where possible, computerized data sets. The selected sites were those from which at least one sample had been collected during 1990-99, and at least one sample of the same species and tissue type had been collected during 1969-89. Once the particular sites, species, and tissue types to be considered were determined, additional data were sought to fill in temporal data gaps. Species and tissue types were treated separately in the analysis.

A preliminary review of available data revealed that the most comprehensive spatial and temporal coverage, with the least spatial and temporal variation in species and tissue types, could be achieved by combining data from the following sources (table 2): (1) the States of New Jersey, New York, and Pennsylvania, (2) three large Federal programs - NSCRS, NCBP, and NAWQA, and (3) a more localized USGS study. Many State monitoring agencies changed from analyzing whole-body fish to analyzing edible portions during the mid 1980's. Additionally, edible-portion samples varied within some species among sites and years as to whether skin was removed or retained on the fillet. These changes were made in order to sample species and tissue that people were likely to consume.



Fish species commonly collected over the years for PCB analyses include white sucker (shown here), common carp, and American eel.

However, these changes confound attempts to make long-term comparisons within species and tissue types for many sites. This problem is compounded by the lack of lipid data for many sites and years, which prevents the use of lipid-normalization to analyze long-term trends in a combined data set (i.e. edible portions and whole fish tissue of various species). Three species were collected most frequently over the years and at the largest number of sites. These were common carp, white sucker, and American eel. The 1990's samples for common carp and white sucker were from whole-body fish; therefore, only whole-body fish data for these species from earlier studies were included in the analysis of data for this report. Edible portions of American eel samples consisted of either muscle sections or the entire fish without head and viscera. A few other species collected during 1994-98 were included to provide information on recent concentrations in edible fish flesh; these were skinless fillets of channel catfish (*Ictalurus punctatus*), and scaled, skin-on fillets of smallmouth bass (*Micropterus dolomieu*).

Data screening and compilation indicated a total of six segments (comprising one or more sites) among the Delaware, Lehigh, and Schuylkill Rivers, and Brandywine Creek (fig. 1) that would be suitable for analysis because each had PCB data from the mid- to late- 1990's, as well as from the previous decade(s), for one or more of the selected fish species and tissue types. These segments are (1) an upper section of the Delaware River, including Upper Knight's Eddy and Port Jervis, N.Y., and Milford and Delaware Water Gap, Pa. (referred to herein as the "upper Delaware River"); (2) a non-tidal portion of the lower Delaware River in the vicinity of Trenton, N.J. and Yardley, Pa. (referred to herein as the "lower Delaware River"); (3) the Lehigh River between Nazareth, Pa. and Easton, Pa., (referred to herein as the "lower Lehigh River"); (4) the Schuylkill River from

sites in the vicinity of Reading, Pa. to a site downstream of Douglassville, Pa. (referred to herein as the "middle Schuylkill River"); (5) the Schuylkill River in the vicinity of Philadelphia, Pa., between Flat Rock Dam and Fairmont Dam (referred to herein as the "lower Schuylkill River"); and (6) Brandywine Creek at Chadds Ford, Pa.

Analysis of temporal patterns in PCB concentrations in fish tissue can be complicated because field and laboratory methods have varied within and among sampling programs with time. Thus, available data are suitable for looking at relatively large differences (e.g. on a logarithmic scale) over time, but are not suitable for examining fine-scale differences. A brief summary of each program's analytical methods, and its method references, are provided in table 2. Most data sets provided total PCB concentration, but the NCBP data set listed concentrations of several Aroclors. As a result, PCB Aroclor concentrations, where given instead of total PCB concentration, were summed to allow comparison of total PCBs among samples. For species and tissue types with multiple reporting limits, the highest value was used as the reporting limit for graphical and (or) statistical analysis. A few samples with unusually high detection limits were eliminated from data sets used for graphical analyses. Censored values (less than the data sets' minimum reporting levels) were set to half the common minimum reporting level for graphical analysis of concentrations. Many river segments and years were represented by a PCB concentration from a single composite sample for a given species and tissue type. For sites at which more than one sample was collected during a given year, the mean concentration was used for plotting in bar charts. Most samples analyzed were composites of three to eight individual adult fish of the same species. A small number of samples consisted of a single specimen.

Temporal trends were examined through graphical analysis, and, where sufficient data points were available, through nonparametric correlation analysis. Concentrations

Table 2. Agencies and programs from which fish-tissue data for selected fish species, tissue types, and sampling sites were obtained for trends analysis of polychlorinated biphenyl (PCB) concentrations in fish from the Delaware River Basin, 1969-98. [American eel (*Anguilla rostrata*) samples are edible flesh; common carp (*Cyprinus carpio*) and white sucker (*Catostomus commersoni*) samples are whole fish; channel catfish (*Ictalurus punctatus*) samples are skinless fillets, and smallmouth bass (*Micropterus dolomieu*) samples are skin-on fillets; $\mu\text{g/g}$, micrograms per gram; mrl, minimum reporting level.]

Agency, program, and years			
Waterbody	Location	Species	Laboratory method, minimum reporting level given, and references
New Jersey Department of Environmental Protection, Toxics in Biota Monitoring Program, 1986-88			
Delaware River	Trenton, N.J.	American eel, channel catfish	Soxhlet extraction with hexane-acetone; clean-up and fractionation with gel-permeation chromatography; characterization by dual-column gas chromatography with electron-capture detection; total PCB concentration quantified by comparison with standards for Aroclors 1248, 1254, and (or) 1260; mrl = 0.1 µg/g (Hauge, 1993; Hauge and others, 1990).
New York State Department of Environmental Conservation, Statewide Toxic Substances Monitoring Program, 1982			
Delaware River	Knights Eddy, N.Y.	American eel, smallmouth bass	Soxhlet extraction with petroleum ether; clean-up with Florisil column; characterization by packed-column gas chromatograph with electron-capture detection; total PCB concentration determined by comparison with standards for Aroclors 1221,1016, and 1254; mrl = 0.1µg/g (Sloan, 1987; Sloan and others, 1983).
Pennsylvania Department of Environmental Protection, Surface-water Quality Monitoring Network , 1976-96			
Brandywine Creek	Chadds Ford, Pa.	American eel, white sucker	1976-1979: analysis by Association of Official Analytical Chemists' multiple residue method; identity and concentration of PCBs confirmed by gas chromatograph-mass spectrometer; mrl = 0.01 - 0.1 µg/g (Brezina, 1980; Brezina and Arnold, 1976). 1980- 1996: Analysis by dual-column gas chromatography with electron-capture detection; total PCB concentration determined by comparison with Aroclor standards; mrl = 0.1 - 0.2 µg/g prior to 1996, and 0.05 µg/g beginning in 1996 (Shertzer and Schreffler, 1996)
Delaware River	Water Gap, Pa. Matamoras, Pa. Milford, Pa. Yardley, Pa.	American eel, white sucker American eel American eel American eel, channel catfish	
Lehigh River	Nazareth, Pa.	white sucker	
Schuylkill River	Reading, Pa. Douglassville, Pa. Philadelphia, Pa.	American eel, channel catfish American eel American eel, common carp, white sucker, smallmouth bass	
U.S. Environmental Protecton Agency, National Study of Chemical Residues in Fish, 1984 and 1986			
Schuylkill River	Philadelphia, Pa.	common carp, white sucker	Extraction with hexane/methylene chloride; fractionation with gel permeation chromatography; characterization with gas chromatography - mass spectrometry with electron-capture detection; PCBs quantified as congeners; mrl =0.00125-0.00625 µg/g, (mrl varies among levels of chlorination, U.S. Environmental Protection Agency, 1992a,b).
U.S. Geological Survey, National Water-Quality Assessment Program, 1998			
Delaware River	Port Jervis, N.Y. Trenton, N.J	white sucker, smallmouth bass common carp, white sucker, smallmouth bass	Soxhlet extraction with methylene chloride; separation with gel permeation chromatography; fractionaton with alumina/silica adsorption chromatography; analysis by dual capillary-column gas chromatography with electron-capture detection; PCBs quantified by comparison with mixed Aroclor standards; mrl = 0.05 µg/g (Crawford and Luoma, 1993; Leiker and others, 1995).
Lehigh River	Easton, Pa	common carp, white sucker, smallmouth bass	
Schuylkill River	Philadelphia, Pa.	common carp, white sucker, smallmouth bass	
U.S. Geological Survey, Schuylkill River Study, 1980			
Schuylkill River	Reading, Pa. Philadelphia, Pa.	American eel American eel, smallmouth bass	Extraction, separation, and analysis according to Association of Official Analytical Chemists' method; mrl = 1 µg/kg (Barker, 1984).
U.S. Fish and Wildlife Service, National Contaminant Biomonitoring Program, 1969-86			
Delaware River	Trenton, N.J.	common carp, white sucker	Methylene chloride extraction; separation by gel permeation chromatography; analysis by gas chromatography with electron-capture detection; total PCB concentration is sum of Aroclors; mrl = 0.1µg/g (Schmitt and others, 1983, 1999).



in fish tissue also were compared with criteria, tolerance levels, and (or) guidelines established or recommended for the protection of human and wildlife health. Some of these criteria, tolerance levels, and (or) guidelines are now many years old, are based on limited toxicity information, and are no longer considered protective. The levels that are protective of human and ecological health are subjects of current research. However, several criteria, tolerance levels and guidelines are included here as points of reference for how concentrations have changed over time. The U.S. Food and Drug Administration (FDA) tolerance level for PCBs (the concentration above which interstate commerce in fish flesh is prohibited) is 2 $\mu\text{g/g}$ in edible flesh (U.S. Food and Drug Administration, 1992); some states use this value as the upper limit beyond which fish should not be consumed at all. The Commonwealth of Pennsylvania uses five consumption advisory groups (Pennsylvania Fish and Boat Commission, 2000) to provide advice to the general population regarding the number of meals (of a particular species from a given water body) to be consumed in a given time period, as follows:

Group	Meal frequency advised	Maximum concentration (micrograms PCB per gram of edible fish flesh)
1	unrestricted	$\leq 0.05^a$
2	1 per week	0.06 - 0.20
3	1 per month	0.21 - 1.0
4	6 per year	1.1 - 1.9
5	none	$> 1.9^b$

a. \leq less than or equal to

b. $>$ greater than

These consumption advisory guidelines are based on the Protocol for a Uniform Great Lakes Sport Fish Consumption Advisory, which was developed by the Great Lakes Fish Advisory Task Force (Anderson and others, 1993).

PCB concentrations in whole fish were compared with 2 levels established for the protection of fish-eating wildlife (1) the New York State Department of Environmental Conservation criterion of 0.11 $\mu\text{g/g}$ (Newell and others, 1987), and (2) the National Academy of Sciences / National Academy of Engineering (NAS/NAE) guideline of 0.5 $\mu\text{g/g}$ (National Academy of Sciences/ National Academy of Engineering, 1973). The NAS/NAE guideline is almost 30 years old, is based on very limited toxicity information, and is set much higher

than concentrations in more recent toxicity-based wildlife guidelines. Although there is no general consensus that concentrations below these levels are safe for consumption by wildlife and fish, they are included in this report as points of reference for how concentrations have changed over time.

In addition to the absence of a general consensus regarding safe levels of total PCBs, none of the criteria or guidelines relating total PCB concentration to human and wildlife health address the relation between potential toxicity and the particular composition of the PCB-congener mixture in a sample (Schmitt and others, 1999). PCB-congener data were not available for most samples included in this study. Thus, temporal trends in the toxicity of samples to humans and wildlife cannot be addressed with the total PCB data used in this study.

PCB CONCENTRATIONS IN FISH TISSUE FROM SELECTED SITES, 1994-98

PCBs were detected in at least one sample of American eel, common carp, white sucker, channel catfish, and (or) smallmouth bass collected during 1994-98 from each of the six river segments (table 3). The lowest concentrations detected were in samples from the upper Delaware River, and the highest were in samples from the middle and lower segments of the Schuylkill River. In general, PCB concentrations were highest in fish samples from sites in urban-industrial watersheds. This is shown in figure 2 by the relation between PCB concentration and human population density. Population density is associated with multiple source conditions such as industrial effluent, sewage treatment plant effluent, as well as runoff and leaching from lands containing electrical transformers and other PCB-containing equipment.

RECENT PCB CONCENTRATIONS IN RELATION TO HUMAN HEALTH CRITERIA

None of the edible-flesh samples collected during 1994-98 exceeded the FDA tolerance level of 2 $\mu\text{g/g}$, but smallmouth bass fillets collected during 1998 from the lower Schuylkill, lower Delaware, and lower Lehigh Rivers exceeded the “unrestricted consumption” level of 0.05 $\mu\text{g/g}$ (Pennsylvania Fish and Boat Commission, 2000), as did channel catfish collected from the middle Schuylkill River in 1995 and American eel collected from Brandywine Creek



“Smallmouth bass collected in 1998 from the lower Schuylkill, Delaware, and Lehigh Rivers exceeded the “unrestricted consumption” level of 0.05 ug/g”.

Table 3. Total polychlorinated biphenyl (PCB) concentrations (wet weight) in edible flesh of American eel (*Anguilla rostrata*), smallmouth bass (*Micropterus dolomieu*), and channel catfish (*Ictalurus punctatus*), and in whole common carp (*Cyprinus carpio*) and white sucker (*Catostomus commersoni*) from selected sites, Delaware River Basin, 1994-98.

[Each entry represents a single composite sample of four to eight individual fish. $\mu\text{g/g}$ ww, micrograms per gram wet weight; na, criterion not directly applicable to tissue sampled; —, criterion less than reporting limit; <, less than; NAWQA, U.S. Geological Survey's National Water-Quality Assessment Program; PADEP, Pennsylvania Department of Environmental Protection. Locations are shown in fig. 1.]

Site	Species	Year	Total PCBs (mg/g ww)	Criterion exceeded				Source
				PA ^a	FDA ^b	NYS ^c	NAS/NAE ^d	
Brandywine Creek								
Chadds Ford, Pa.	American eel	1994	0.58	YES	NO	na	na	PADEP
Upper Delaware River								
Milford, Pa.	American eel	1995	<.36	—	NO	na	na	PADEP
Port Jervis, NY/Milford, Pa.	American eel	1995	<.37	—	NO	na	na	PADEP
	smallmouth bass	1998	<.05	NO	NO	na	na	NAWQA
	white sucker	1998	.14	na	na	YES	NO	NAWQA
Water Gap, Pa.	American eel	1994	.27	YES	NO	na	na	PADEP
	American eel	1995	<.34	—	NO	na	na	PADEP
	American eel	1996	.05	NO	NO	na	na	PADEP
Lower Delaware River								
Trenton, NJ / Yardley, Pa.	American eel	1995	<.26	—	NO	na	na	PADEP
	smallmouth bass	1998	.37	YES	NO	na	na	NAWQA
	common carp	1998	1.10	na	na	YES	YES	NAWQA
	white sucker	1998	.26	na	na	YES	NO	NAWQA
Lower Lehigh River								
Easton, Pa.	smallmouth bass	1998	.08	YES	NO	na	na	NAWQA
	common carp	1998	.9	na	na	YES	YES	NAWQA
	white sucker	1998	.64	na	na	YES	YES	NAWQA
Schuylkill River, Reading								
Reading, Pa.	channel catfish	1995	1.3	YES	NO	na	na	PADEP
Lower Schuylkill River								
Philadelphia, Pa.	smallmouth bass	1998	.25	YES	NO	na	na	NAWQA
	common carp	1998	4.0	na	na	YES	YES	NAWQA
	white sucker	1998	.89	na	na	YES	YES	NAWQA

^a Pennsylvania Fish and Boat Commission (2000); consumption advisories for the general population, in micrograms per gram wet weight, edible flesh:

1 = unrestricted, 0 - 0.05

2 = 1 meal per week, 0.06 - 0.20

3 = 1 meal per month, 0.21 - 1.0

4 = 6 meals per year, 1.1 - 1.9

5 = no consumption, greater than 1.9

^b FDA, U.S. food and Drug Administration tolerance level (1992); 2 $\mu\text{g/g}$ wet weight, edible flesh

^c NAS/NAE, National Academy of Sciences and National Academy of Engineering wildlife guideline level (1973); 0.11 $\mu\text{g/g}$ wet weight, whole fish

at Chadds Ford in 1994. An American eel sample collected from the upper Delaware River in 1994 exceeded the “unrestricted consumption” level, but the American eel sample collected from the same section in 1996 did not exceed that level (table 3).

RECENT PCB CONCENTRATIONS IN RELATION TO WILDLIFE CRITERIA

The NYSDEC wildlife criterion ($0.11 \mu\text{g/g}$) was exceeded in whole-body samples of common carp and white sucker collected from the four sites sampled in 1998, in muscle tissue of American eel collected from Brandywine Creek in 1994, and in channel catfish collected from the middle Schuylkill River in 1995. The higher level of $0.5 \mu\text{g/g}$ specified by the older NAS/NAE guideline also was exceeded in whole-body common carp and white sucker samples collected from the lower Lehigh and Schuylkill Rivers in 1998, in the channel catfish fillet sample collected from the middle Schuylkill River in 1995, and in the American eel sample collected from Brandywine Creek in 1994. The NAS/NAE guideline was exceeded by concentrations in carp (but not in white sucker) collected from the lower Delaware River in 1998. This guideline was not exceeded by concentrations in white sucker collected from the upper Delaware River in 1998 (table 3).

TEMPORAL TRENDS IN PCB CONCENTRATIONS IN FISH TISSUE FROM SELECTED SITES, 1969-98

Among river segments considered in this study, the lower Delaware River segment was represented in the data set by the largest number of years (17) during which samples were collected, and spanned the longest time period - from 1969-98. The other rivers and segments had sample data for 6 - 11 years spanning 1976-95 (middle Schuylkill River), 1977-94 (Brandywine Creek), 1979-98 (lower Lehigh River), and 1980-98 (upper Delaware River and lower Schuylkill Rivers).

LOWER DELAWARE RIVER, 1969-98

PCBs were detected in all species and tissue types during most years in which samples were collected from the lower Delaware River. Exceptions were American eel samples collected during 1977, 1993, and 1995 (fig. 3), in

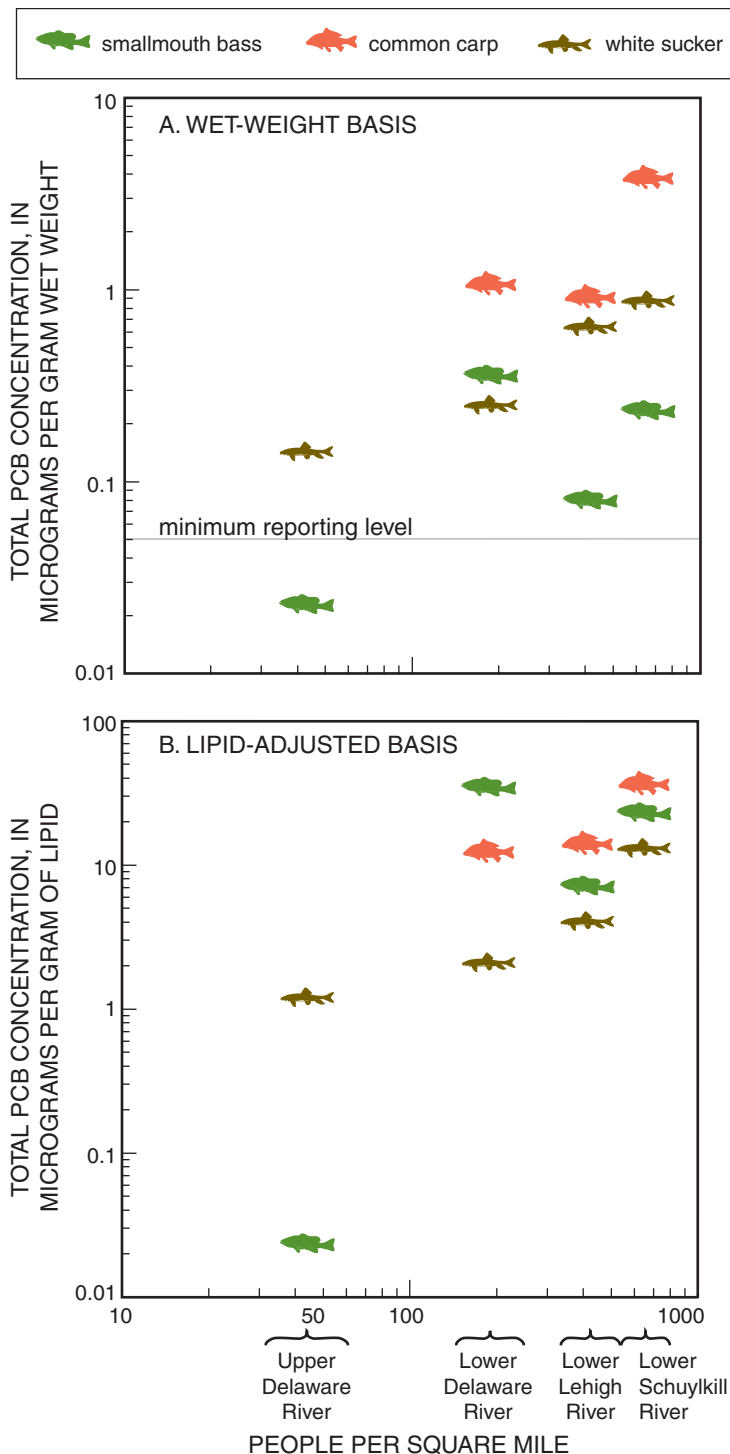


Figure 2. Polychlorinated biphenyl (PCB) concentrations in fish tissue collected in 1998, in relation to 1990 population densities at four river segments in the Delaware River Basin. Concentrations are shown on (A) wet-weight basis and (B) lipid-adjusted basis. Each point represents a composite sample of 3-8 adult fish. Smallmouth bass (*Micropterus dolomieu*) samples were composed of scaled, skin-on fillets; common carp (*Cyprinus carpio*) and white sucker (*Catostomus commersoni*) samples were composed of whole fish.



(Photograph by David B. Soete, Honesdale, Pa.)

which PCBs were not detected (one 1977 sample and both 1990's samples) or were detected at a concentration less than the lowest common reporting limit ($0.26 \mu\text{g/g}$) used for American eel data in figure 3 (the other 1977 sample).

A decline in PCB concentrations is evident for most species collected from the lower Delaware River during 1969-98 (fig. 3). The correlation between PCB concentration in whole white sucker and year was statistically significant. Spearman rank analysis resulted in a correlation of -0.46 ($p = 0.08$, $n=15$). This correlation improved (-0.53 , $p = 0.05$, $n = 14$) when a 1977 sample with an unusually low concentration ($0.1 \mu\text{g/g}$) relative to other samples collected during the mid-1970's was eliminated from the analysis. PCB concentrations in American eel declined during this time period, from a highest concentration of $3.8 \mu\text{g/g}$ (wet weight) in a sample collected during 1976 to less than the detection limit of $0.26 \mu\text{g/g}$ (wet weight) in a sample collected during 1995 (fig. 3). However, the decline is not statistically significant due to the low or non-detected concentrations in the two samples from 1977. Concentrations in most species declined from above the relatively high NAS/NAE wildlife-protective guideline level ($0.5 \mu\text{g/g}$) in the 1970's to below this level in the 1990's. However, the PCB concentration in a common carp sample collected during 1998 remained above the NAS/NAE guideline level, and the PCB concentration in a whole white sucker sample collected during 1998 exceeded the NYSDEC wildlife criterion ($0.11 \mu\text{g/g}$). PCB concentrations in edible flesh of American eel and channel catfish from the lower Delaware

River exceeded the FDA tolerance level ($2 \mu\text{g/g}$) in the 1970's (American eel) and 1980's (channel catfish), and dropped to below this tolerance level in the mid- 1990's. A general decline in all species studied, except carp and channel catfish, can be seen in the plot of concentrations calculated on a lipid-adjusted basis (fig. 3B). The decline in lipid-adjusted PCB concentrations in whole white sucker samples was statistically significant (Spearman correlation = -0.72 , $p < 0.01$, $n = 15$).

ALL SITES, 1976-98

Polychlorinated biphenyl data were available for whole-body white sucker samples collected during 1979-87 as well as during 1998 from the upper Delaware, lower Delaware, lower Lehigh, and lower Schuylkill Rivers. PCB data were also available for whole-body common carp samples collected during the 1980's as well as during 1998 from the lower Delaware and Schuylkill Rivers. Data were available for American eel (edible flesh) samples collected during the 1980's as well as during the early- to- mid-1990's, from both Delaware River segments, both Schuylkill River segments, and from Brandywine Creek. American eel data from the mid-1970's were also available for the middle Schuylkill River and the lower Delaware River.

Whole-body White Sucker

PCB concentrations in whole-body white sucker samples collected during 1998 were highest in those collected from the lower Schuylkill River, followed by the lower Lehigh River (fig. 4A). Concentrations were lowest in the sample from the upper Delaware River. PCB concentrations in fish from the upper Delaware River and from Brandywine Creek show relatively large initial declines in the early 1980's, followed by fairly stable concentrations in subsequent samples (fig. 4A). The absence of whole white sucker data after 1985 for Brandywine Creek does not permit examination of more recent concentrations to determine if the decline has continued for this species. The lower Delaware River, which had the highest PCB concentrations in white sucker samples collected during the early 1980's, exhibited the most dramatic improvement of the rivers studied, and has shown a clear decline in PCB concentrations over time.

Trends in PCB concentrations in whole-body white sucker samples collected from the lower Schuylkill and Lehigh Rivers were more difficult to interpret than those

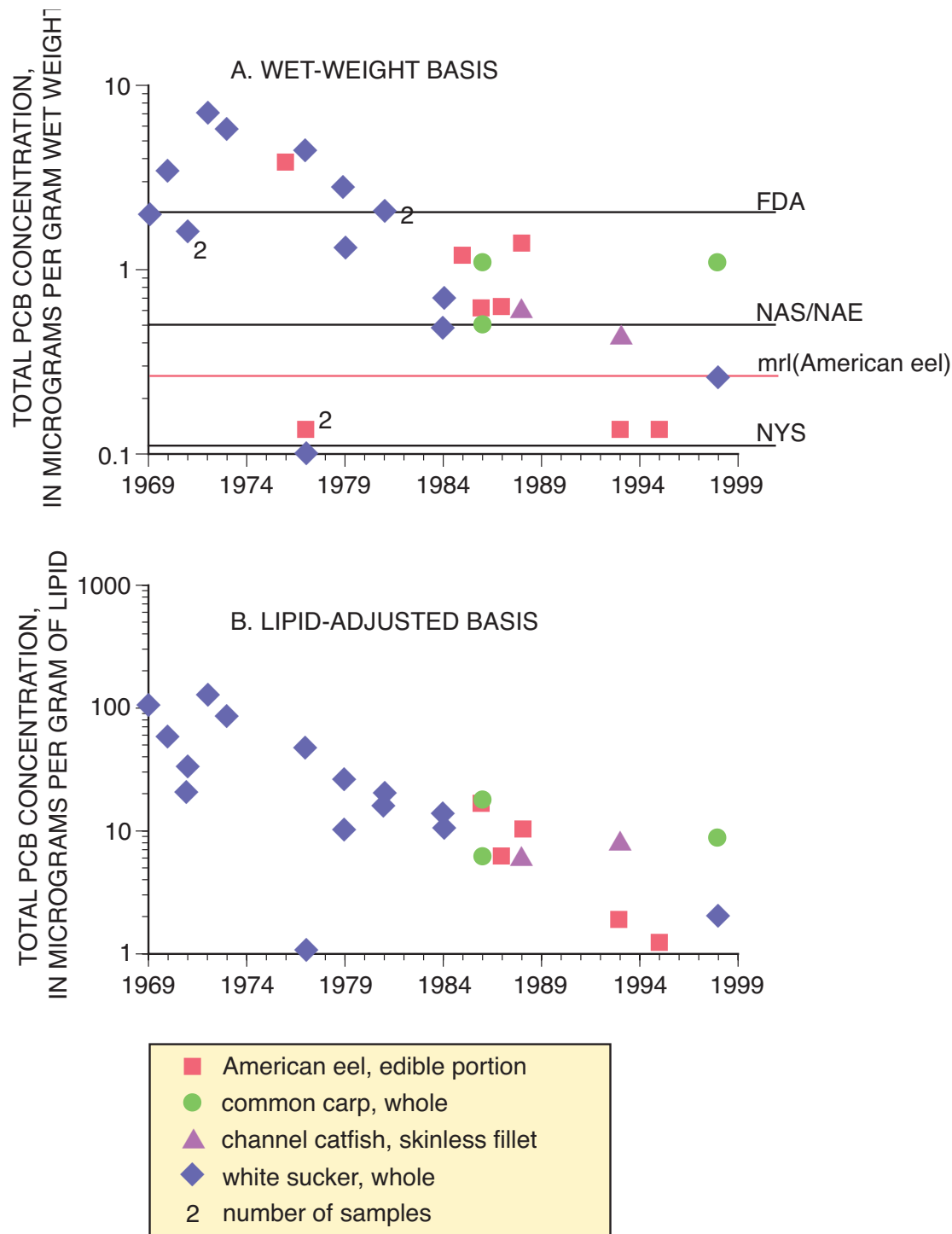


Figure 3. Temporal trends in polychlorinated biphenyl (PCB) concentrations in fish from the lower Delaware River, 1969-98, shown on (A) wet-weight basis and (B) lipid-adjusted basis. [American eel, *Anguilla rostrata*; common carp, *Cyprinus carpio*; channel catfish, *Ictalurus punctatus*; white sucker, *Catostomus commersoni*; FDA, U.S. Food and Drug Administration tolerance level; NAS/NAE, National Academy of Sciences and National Academy of Engineering wildlife guideline level (1973); NYS, wildlife protection criterion used by New York State Department of Environmental Conservation; mrl, minimum reporting level for American eel samples].



“PCB concentrations in fish collected from the Lower Schuylkill River segment during 1998 were above the national geometric mean concentration reported for 1979”.

for other sites. PCB concentrations in samples collected from the lower Lehigh River in 1998 (wet-weight basis) were about the same as in 1979. An initial decrease after 1979 was followed by a general pattern of increasing concentration with time. In contrast, the 2 samples from this site for which lipid data were available actually show a decrease in lipid-adjusted PCB concentrations for this time period (fig. 4B). This suggests differences in fat content may be obscuring a decline in PCB concentrations in samples from the lower Lehigh River. Similarly, PCB concentrations in the white sucker sample collected from the lower Schuylkill River during 1998 indicate a minor and inconsistent decrease since the early 1980's, but an increase over the 1987 concentration. The cause of this inconsistency also may be variation in lipid content. The lipid-adjusted PCB concentrations in the two samples actually show a decrease between the two years (fig. 4B).

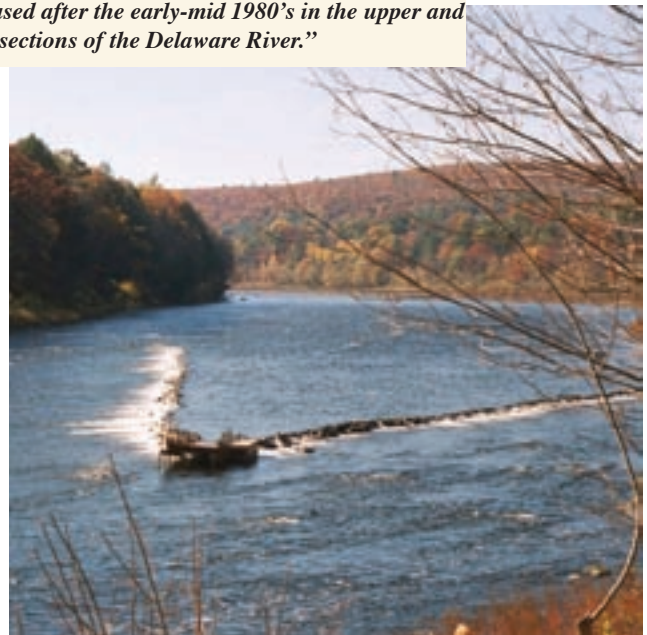
Edible flesh of American Eel

PCB concentrations in edible flesh of American eel (fig. 5) show trends that are similar to those of whole-body white sucker—decreasing over time in the upper and lower Delaware River segments, and showing no clear trend in the lower Schuylkill River, where concentrations in 1992 still exceeded the FDA tolerance level. PCB concentrations in American eel samples from the middle Schuylkill River declined between the mid-late 1970's and the mid 1980's, but a sample collected during the early 1990's shows no further change. The upper and lower Delaware River segments showed the most dramatic decline of all 5 segments for which American eel data were available. PCB data for American eel samples collected from Brandywine Creek in 1995 indicate a continuation of the decline seen in whole-body white sucker samples from this site (fig. 4A).

Whole-body Common Carp

PCB concentrations in whole-body common carp collected during 1998 from the lower Delaware, lower Schuylkill, and lower Lehigh Rivers remained above levels considered protective of wildlife, and were higher than the 1979 National geometric mean (Schmitt and others, 1990). PCB concentrations in whole-body common carp collected during 1998 were highest in the lower Schuylkill River sample. Concentrations were similar between the lower Delaware and Lehigh Rivers (fig 4A, 4B). Plots of wet weight and lipid adjusted PCB concentrations in whole-body common carp from the 1980's to 1998 show minimal change at the lower Delaware River and increases in the lower Schuylkill River (figs. 4A, 4B). No common carp trend data were available for the lower Lehigh river.

“PCB concentrations in edible flesh of American eel decreased after the early-mid 1980's in the upper and lower sections of the Delaware River.”



(Photograph of eel weir by David B. Soete, Honesdale, Pa.)

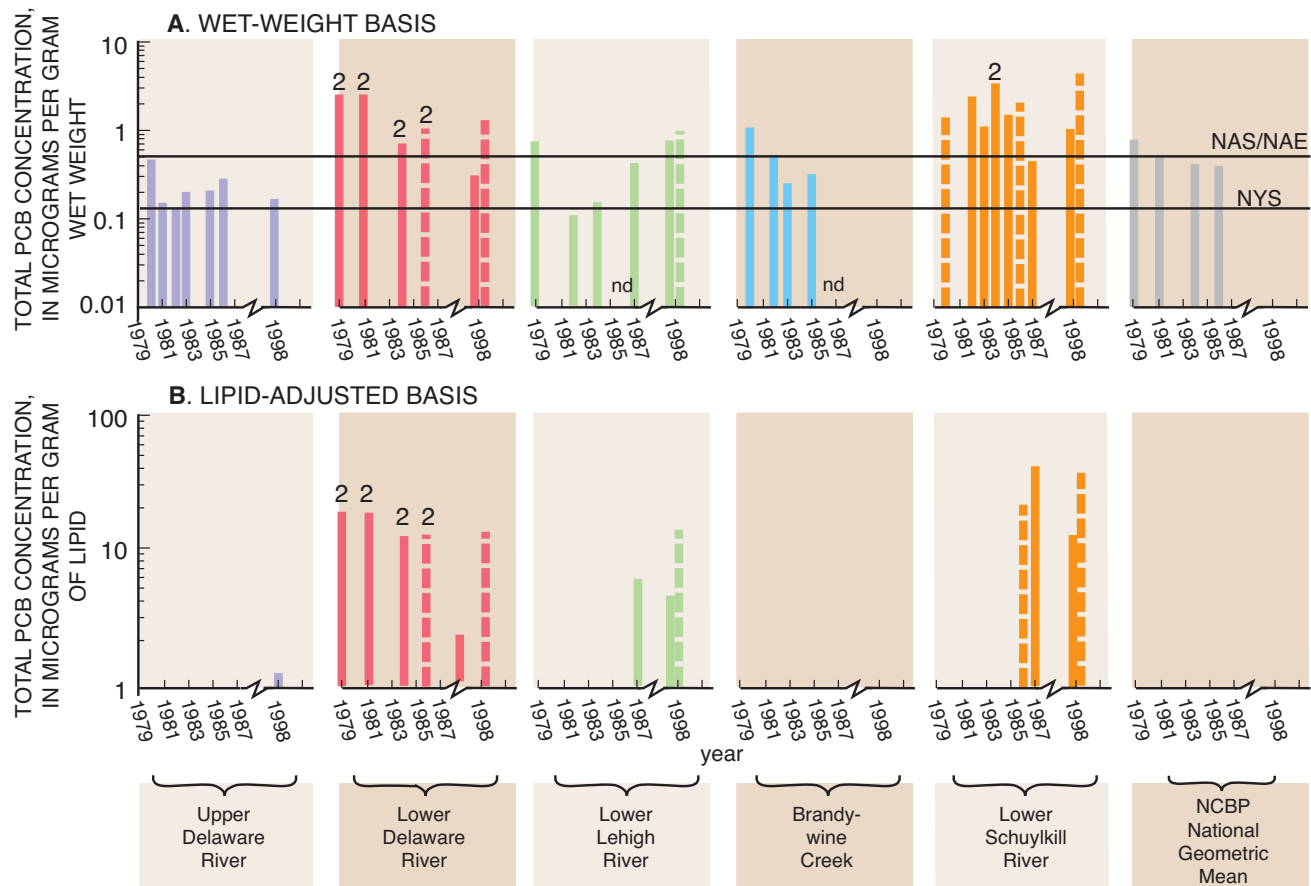


Figure 4. Polychlorinated biphenyl (PCB) concentrations in whole white sucker (*Catostomus commersoni*, solid bars) and common carp (*Cyprinus carpio*, hatched bars) samples from selected river segments in the Delaware River Basin, 1979-98. Concentrations are shown on (A) wet-weight basis and (B) lipid-adjusted basis. Site locations are shown in figure 1. National geometric mean concentrations (1979-86) from the National Contaminant Biomonitoring Program (NCBP) are also shown. Bars represent the concentration from a single composite sample, or (where indicated by a number above the bar) the mean concentration of multiple composite samples. [NAS/NAE, wildlife guideline level recommended in 1973 by the National Academy of Sciences and National Academy of Engineering; NYS, wildlife protection criterion used by New York State; nd, not detected at reporting level of 0.2 micrograms per gram].

FACTORS THAT AFFECT TRENDS IN PCB CONCENTRATIONS IN FISH TISSUE

Trends in PCB concentrations in fish tissue were quite variable among the six sites examined. The upper and lower Delaware River segments show considerable improvement, the upper Schuylkill and the Brandywine Creek sites show steady declines, and the lower Schuylkill and Lehigh sites have more ambiguous trend data. The general decline at most sites indicate PCBs are less available than in the past. Site-to-site differences in the rate of PCB decline, and in the resulting concentrations in the 1990's, are probably due to differences in (1) the proximity, extent, severity, and longevity of PCB

sources, (2) hydrologic factors that affect the movement and retention of contaminated sediment, and (3) habitat features that affect the duration of exposure of resident fish to contaminated sediments. For instance, PCBs in white sucker and American eel from the upper Delaware River, which is the least populated and industrialized of the river segments considered in this study, declined to fairly low levels. The detection of PCBs in fish from the less-industrialized upper Delaware River segment is not unexpected; PCBs can be transported within and among river basins through atmospheric transport, relocation of contaminated sediment, and disposal of equipment containing PCBs (Schmitt and others, 1983). Older contaminated sediments that can be resuspended in storms

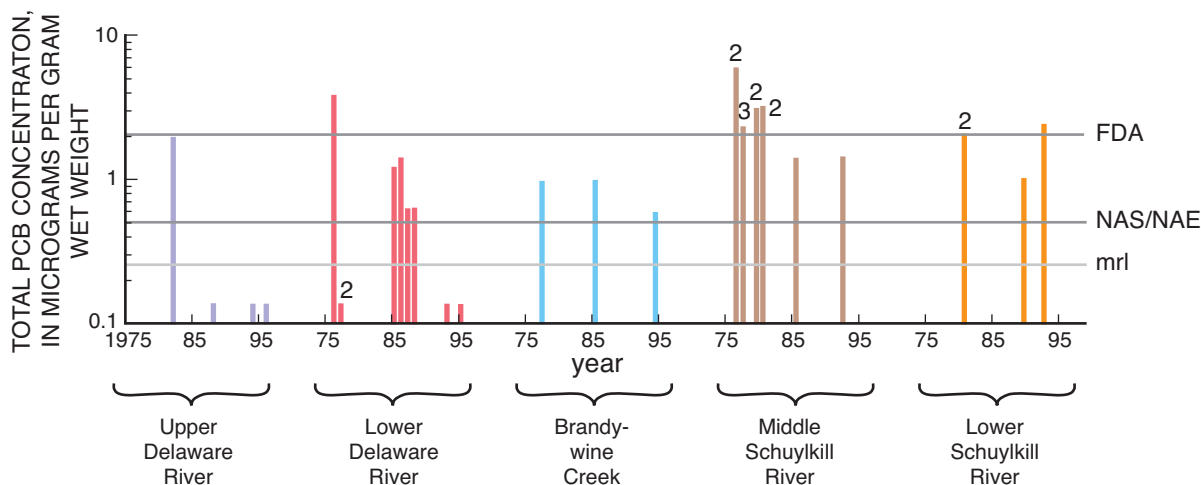


Figure 5. Polychlorinated biphenyl (PCB) concentrations in samples of edible flesh portion of American eel (*Anguilla rostrata*) from selected river segments in the Delaware River Basin, 1976-95. Site locations are shown in figure 1. Each bar represents a single composite sample, or (where indicated with number above the bar) the mean of multiple composite samples. [FDA, U.S. Food and Drug Administration tolerance level; NAS/NAE, wildlife-protection criterion recommended by National Academy of Sciences and National Academy of Engineering in 1973; mrl, minimum reporting level common to all samples].

can also be a source. The fact that PCBs are still detected and have shown little decline in the past few years, both here and at other sites, probably indicates they are still being transported.

Polychlorinated biphenyl concentrations in white sucker from the lower Delaware River have shown a relatively clear decline over the years, while the lower Schuylkill and Lehigh Rivers have not shown consistent declines. The decline on the lower Delaware is especially significant given that the lower Delaware and lower Schuylkill Rivers had similar levels of contamination in the early 1980's. Variation in lipid concentration might be partly responsible for the lack of clear patterns in decline at the lower Schuylkill and Lehigh Rivers. Both sites show a decline when viewed on a lipid basis, but lipid data are too few to be conclusive. Some other factors that might account, in part, for the lack of a clear decline could include differences in age, size, gender, and habitat, as well as differences in environmental concentrations and sources of PCBs. Another factor may be the dams and pools from which fish were collected on the lower Schuylkill and Lehigh Rivers. Dams could contribute to the relatively high concentrations in fish tissue by trapping contaminated sediment and providing a longer exposure to fish than in free-running rivers. Similar findings have been reported in the upper Mississippi River, with its many large pools (Lee and Anderson, 1998) and in some New England rivers with many dams (Breault and Harris, 1997). Despite the lack of sufficient samples to establish a clear trend

for the lower Schuylkill River, the data indicate that this river segment has undergone the least improvement of the six river segments studied. PCB concentrations in fish from both of the Schuylkill River segments and the lower Lehigh River remain at high levels during the 1990's relative to (1) the other rivers considered in this study, (2) human-health and (or) wildlife protective guidelines, and (3) national geometric means from almost two decades prior. High population density and industrialization in these watersheds is closely related to PCB contamination. These findings are consistent with results of NAWQA studies conducted during 1992-95 that compared PCB concentrations in white sucker and (or) common carp among sites in various land-use settings within major river basins throughout the Nation. For example, NAWQA studies of numerous rivers and streams within the Hudson River Basin (Wall and others, 1995), the Connecticut River Basin (Coles, 1998), and the Lower Susquehanna River Basin (Bilger and others, 1999) all found higher PCB concentrations in fish from rivers and streams draining urban-industrial centers than in fish from rivers and streams draining watersheds with other land uses.

Streambed-sediment samples collected during 1998 as part of the Delaware River Basin NAWQA are consistent with the fish-tissue results collected the same year and presented here. PCB concentrations in bottom sediment from the Schuylkill and Lehigh Rivers (0.085 and 0.15 µg/g, respectively) in 1998 were higher than in those from the upper and lower Delaware River (both less than

the detection limit of $0.05 \mu\text{g/g}$; DeLuca and others, 1999). PCB concentrations ($0.14 \mu\text{g/g}$) in stream-bed sediment from the Schuylkill River at Berne, about 15 mi. upstream from Reading, also were higher than in bed sediment at other sites; this result indicates a relatively wide distribution of PCBs in the Schuylkill River.

PCB concentrations in whole-body common carp collected during 1998 at each site typically were higher than in other species collected that year from the same sites. Differences among species in PCB concentrations can be affected by interspecific variation in fat content, behavior, age, size, habitat, feeding, and sexual condition. Common carp show a lack of change in PCB concentrations over the years, and (at some sites), even a possible increase where other species show a decrease. The identification of trends for this species is limited by the small number of whole-body common carp samples (1 - 3 for any site) available for this study.

The variety of species and tissue types among sites and years, and the representation of many sites and years by single or few samples, makes the analysis of temporal patterns in PCB concentrations at many sites difficult. Future analysis of temporal trends would be enhanced by collection of the same species and tissue type at selected sites, and collection of multiple samples of particular species so that variance can be assessed.

SUMMARY AND CONCLUSIONS

Trends in PCB concentrations in fish tissue collected from selected rivers in the Delaware River Basin during 1969-98 were investigated as part of the U.S. Geological Survey's NAWQA Program. Data compiled from State and Federal programs in which fish were collected for PCB analyses during 1969-1996 were combined with NAWQA data collected during 1998. The numbers of species, tissue types, and rivers chosen for this study were sharply limited so as to facilitate comparisons among sites and years. Data on whole white sucker, whole common carp, and edible flesh of American eel were used to examine temporal trends at the upper Delaware River, the lower Delaware River, the middle Schuylkill River, the lower Schuylkill River, the lower Lehigh River, and Brandywine Creek at Chadds Ford, Pa. Additional data on edible flesh of channel catfish and smallmouth bass were included to identify recent (1994-98) concentrations and relate them

The lower Schuylkill River has undergone the least improvement in fish-tissue PCB concentrations of the six river segments studied.



(Photograph by Nathan Gasser, Philadelphia, Pa.)

to human-health and wildlife-health levels. Trends were examined by graphical and correlation analyses, and concentrations were compared with selected human-health and wildlife criteria and (or) guidelines.

PCB concentrations in edible and whole-body fish tissue of several species were lowest most years in fish from the upper Delaware River, where concentrations in edible flesh of game fishes have decreased in the past 20 years to below the levels at which fish consumption advisories are established by Pennsylvania. However, concentrations in a whole white sucker sample collected during 1998 still exceed the NYSDEC wildlife criterion concentration and may pose a risk to fish-eating wildlife.

Four of the six river segments have shown a decline in the concentration of PCBs in whole white suckers and (or) edible portions of American eel since the 1970's and (or) early 1980's. This trend is especially apparent in the lower Delaware River, where PCB concentrations in edible fish tissue and whole-body fish tissue from this site have declined by an order of magnitude and dropped from above the FDA tolerance level and the relatively high NAS/NAE wildlife guideline in the 1970's and 1980's to below these levels in the 1990's. Yet, concentrations in a sample of edible portions of smallmouth bass and a sample of whole white suckers collected from the lower Delaware River during 1998 indicate that PCB concentrations still may pose a risk to human health and wildlife in this section of the Delaware River. White sucker data from Brandywine Creek at Chadds Ford indicate reductions in PCB concentrations between the early 1980's and the mid-1980's, and American eel data indicate a decrease between 1985 and 1995. PCB concentrations in American eel from the middle Schuylkill River, where some of the highest concentrations were reported in the 1970's, have declined, but still exceeded

fish-consumption advisory levels and wildlife-protection criteria in the mid-1990's.

The small sample size, and the limited number of species and tissue types obtained from individual sites, and the variety of species and tissue types preclude a comprehensive analysis of PCB trends among sites and years. Available data, however, indicate recent (1994-98) PCB concentrations in whole white sucker and edible flesh of American eel in the upper Delaware River, the lower Delaware River, the middle Schuylkill River, and Brandywine Creek are lower than in the 1970's and (or) 1980's. A decline in PCB concentrations is not as apparent in some species collected from the lower Lehigh and lower Schuylkill Rivers. PCB concentrations in fish from the lower Lehigh river and the lower Schuylkill River have decreased at a much slower rate than in fish from the other rivers and river segments considered in this study. Why some rivers have improved more than others is uncertain. Despite observed declines at some of these sites, PCB concentrations in one or both whole-body fish samples collected in 1998 from the lower Schuylkill, lower Lehigh, and lower Delaware Rivers exceeded the national geometric mean concentration from nearly 2 decades earlier; these elevated concentrations seem to be related to degree of industrialization and urbanization of these watersheds. More information on the composition of the PCB mixtures would be necessary to evaluate the potential toxicity of the PCBs present in these samples. However, the PCB concentrations in some fish species collected in recent years from most of these river segments remain at levels of concern for human and wildlife health.

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(Photograph by Barry P. Baldigo, USGS)

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